# Appendix C.  Key Radionuclides and Generation Processes

This appendix presents the criteria that was used to select the key radionuclides that were analyzed in this Report. It contains a more complete discussion of the major key radionuclides that have the greatest impact on DOE LLW disposal capacity. A discussion of the major processes that generate DOE LLW is provided, and finally, a discussion of Special Case Waste is provided.

## C.1    Criteria for Selection of Radionuclides:

The following criteria were developed to select the list of radionuclides that are important to evaluation of radiological capacity at DOE LLW disposal facilities:

- Half-life in excess of 5 years.

- Identified in site documents from throughout the DOE complex as a radionuclide critical to evaluation of dose to the public through ground water, atmospheric, or inadvertent intruder scenarios.

- Daughter products of the identified radionuclides are excluded from the list. Since doses from radiologically significant daughter products are considered in the estimation of parent nuclide dose (equilibrium stage), the activity of the daughters need not be reported. Reporting both parents and daughters can be confusing and may lead to an overestimate of the radionuclide inventory and resultant dose.

- Categories for C-14 and Ni-63 activated metal were added because the analysis of radiological capacity recognizes a lower leach rate for radionuclides within a metal matrix.

- Categories for "Uranium-Natural" and "Plutonium-Weapons" were added to facilitate user identification of nuclide profiles.

These criteria resulted in 51 nuclides being included in the 1997 Technical Data Request. Table C-1 provides the list of nuclides considered.

## Table C-1.  Key Radionuclides

| Radionuclide | Abbreviated Form | Atomic Number | Half-Life | Radionuclide | Abbreviated Form | Atomic Number | Half-Life |
|---|---|---|---|---|---|---|---|
| aluminum 26 | Al-26 | 13 | $7.4 \times 10^5$ y | plutonium 238 | Pu-238 | 94 | 86.4 y |
| americium 241 | Am-241 | 95 | 458 y | plutonium 239 | Pu-239 | 94 | 24.39 y |
| americium 243 | Am-243 | 95 | $7.95 \times 10^3$ y | plutonium 240 | Pu-240 | 94 | 6580 y |
| barium 133 | Ba-133 | 56 | 7.2 y | plutonium 241 | Pu-241 | 94 | 13.2 y |
| carbon 14 | C-14 | 6 | 5730 y | plutonium 242 | Pu-242 | 94 | $3.79 \times 10^5$ y |
| carbon 14[*] | C-14 am | 6 | 5730 y | plutonium 244 | Pu-244 | 94 | $7.6 \times 10^5$ y |
| cadmium 113[**] | Cd-113m | 48 | 13.6 y | radium 226 | Ra-226 | 88 | 1602 y |
| chlorine 36 | Cl-36 | 17 | $3.1 \times 10^5$ y | radium 228 | Ra-228 | 88 | 6.7 y |
| curium 243 | Cm-243 | 96 | 32 y | selenium 79 | Se-79 | 34 | $6.5 \times 10^4$ y |
| curium 244 | Cm-244 | 96 | 17.6 y | samarium 151 | Sm-151 | 62 | 87 y |
| cobalt 60 | Co-60 | 27 | 5.26 y | tin 121[**] | Sn-121m | 50 | 76 y |
| cesium 135 | Cs-135 | 55 | $3.0 \times 10^6$ y | tin 126 | Sn-126 | 50 | $10^5$ y |
| cesium 137 | Cs-137 | 55 | 30.0 y | strontium 90 | Sr-90 | 38 | 28.1 y |
| europium 152 | Eu-152 | 63 | 12 y | technetium 99 | Tc-99 | 43 | $2.12 \times 10^5$ y |
| europium 154 | Eu-154 | 63 | 16 y | thorium 229 | Th-229 | 90 | 7340 y |
| tritium | H-3 | 1 | 12.3 y | thorium 230 | Th-230 | 90 | $8.0 \times 10^4$ y |
| iodine 129 | I-129 | 53 | $1.7 \times 10^7$ y | thorium 232 | Th-232 | 90 | $1.4 \times 10^{10}$ y |
| potassium 40 | K-40 | 19 | $1.26 \times 10^9$ y | uranium 232 | U-232 | 92 | 72 y |
| niobium 93[**] | Nb-93m | 41 | 13.6 y | uranium 233 | U-233 | 92 | $1.62 \times 10^5$ y |
| niobium 94 | Nb-94 | 41 | $2.0 \times 10^4$ y | uranium 234 | U-234 | 92 | $2.47 \times 10^5$ y |
| nickel 59 | Ni-59 | 28 | $8 \times 10^4$ y | uranium 235 | U-235 | 92 | $7.1 \times 10^8$ y |
| nickel 63 | Ni-63 | 28 | 92 y | uranium 236 | U-236 | 92 | $2.39 \times 10^7$ y |
| nickel 63[*] | Ni-63 am | 28 | 92 y | uranium 238 | U-238 | 92 | $4.51 \times 10^9$ y |
| neptunium 237 | Np-237 | 93 | $2.14 \times 10^6$ y | natural uranium | U-nat | 92 | N/A |
| protactinium 231 | Pa-231 | 91 | $3.25 \times 10^4$ y | zirconium 93 | Zr-93 | 40 | $1.5 \times 10^6$ y |
| weapons-grade plutonium | Pu Weap | 94 | N/A | | | | |

[*]Activated metal; [**]Metastable

The following nuclides were reported to be below 1 curie throughout the DOE complex: Al-26, Cd-113m, Cm-243, Pa-231, Pu-244, Ra-228, Se-79, and Th-229.

## C.2   Key Radionuclides

This section provides additional detail on the characteristics and sources of the radionuclides that are most important for assessing radiological impacts on Department disposal operations.

### C.2.1   Tritium

Tritium ($^3$H) is a radioactive isotope of hydrogen.  It has a half-life of 12.3 years and decays to helium-3 ($^3$He) with the emission of a beta particle and no gamma ray.  Tritium produces a very low energy beta particle and is usually considered one of the least radiologically hazardous of the

radionuclides. However, since it can replace normal hydrogen in chemical compounds essential for life, tritium poses a potential hazard that can be very mobile within the biological environment.

Light water reactors produce a small amount of tritium, primarily from ternary fission. Most tritium created in these reactors remains in the fuel rods. The major source of tritium in the DOE complex was the tritium production reactors. None of these reactors are currently producing, but a stockpile of tritium remains as part of the DOE mission. Presently, tritium is used in testing, component maintenance, and research applications.

The primary chemical waste forms of tritium from DOE activities are tritium gas and tritiated water, either as a liquid or vapor. Additionally, tritium waste forms generated from the thermonuclear weapons program (e.g., tritium production reactors) are lithium metal hydrides.

Tritium substitutes readily for ordinary hydrogen in water, and thus becomes part of the hydrological cycle. Through sandy soils, migration of tritium takes place at the same velocity as the ground water through sandy soils. However, the migration of tritium in ground water is influenced by the soil type. Tritium migration is not slowed in sandy soils, but in clay soils, a small number of tritium atoms may exchange with ordinary hydrogen atoms in water molecules that are bound within the clay, thus slowing the migration.

The mechanisms of tritium gas uptake in vegetation are not well understood, but plants have the capability to oxidize tritium to form tritiated water. Many of the metabolic reactions that take place in living organisms are involved with the transfer of hydrogen. Any of these reactions can involve tritium instead of ordinary hydrogen. The oxygen and nitrogen atoms in organic molecules make it much easier for tritium to replace hydrogen associated with these groups than with hydrogen attached to carbon atoms.

The biological half-life of tritium in the human body is about 10 to 12 days. The uptake of hydrogen in humans can occur through inhalation, ingestion, or absorption through the skin. The rate of inhalation is about two times the rate of absorption through the skin.

## C.2.2 Cesium-137

Cesium-137 ($^{137}$Cs) is not a naturally occurring radionuclide. Its half-life is 30.2 years, and it emits one to two high-energy beta particles. Approximately 85 percent of all $^{137}$Cs decays result in the emission of a 662 keV gamma ray. Due to the gamma ray energy of $^{137}$Cs, it has been used to sterilize medical supplies, and milk cartons, and irradiate food.

The primary means of producing $^{137}$Cs is via nuclear fission. Industrial applications of $^{137}$Cs include the production of plastic shrink tubing (irradiated plastic has the tendency to shrink after being heated); radiography to inspect metal castings and welds for flaws and material defects (e.g., cracks in steel pipes); radioactive measurement gauges for liquid or solid thicknesses (e.g., gaging of automobile sheet steel); treatment of sewage sludge to kill bacteria and viruses; and medical radiotherapy to kill cancerous tissue.

Chemically, cesium behaves like other alkali metals (lithium, sodium, potassium, etc.) and has the simplest of elemental chemistries (excepting the inert gases, such as helium). Cesium reacts with most other elements directly and reacts explosively with air and water. Therefore, metallic cesium

must be handled in an inert atmosphere. Cesium reacts with most nonmetals to produce one or more binary compounds; it also forms numerous alloys and compounds with other metals, such as lead and tin. Various oxides, sulfides, and similar compounds are readily hydrolyzed by water-forming cesium hydroxides. Cesium salts are generally characterized by high melting points, electrical conductivity of the melts, and high solubility in water.

The largest source of $^{137}$Cs, and therefore the largest potential for waste material, is from nuclear reactors and the large inventory of $^{137}$Cs stored in the irradiated fuel. DOE also maintains a large inventory of $^{137}$Cs as Special Case Waste in the form of sealed sources. These sources were designed to generate known amounts of gamma radiation for equipment and food sterilization, as well as other purposes. A small quantity of $^{137}$Cs waste is also generated in research facilities. The primary waste forms that may contain $^{137}$Cs, in addition to other fission products, include contaminated scrub water, decontamination solutions, demineralizers, contaminated clothing or gloves, contaminated tools and equipment, ion exchange resins, and filters.

The behavior of cesium in the environment is dictated by its chemical properties. Because of its high solubility, cesium generally moves with the ground water but is retained in clay soils. If cesium is not held in the soils, it can relocate through the root system of plants or through the leaves and stems by atmospheric deposition. Cesium-137 deposited on leaves is likely to penetrate into the plant more rapidly than cesium absorbed from the ground.

Absorption of cesium by humans takes place primarily through the digestive tract. Cesium, like potassium, enters body cells; however, cesium is bound more tightly than potassium. The biological half-life for cesium in humans depends on age, sex, and body mass. For adult humans, the biological half-life varies between 50 and 150 days, with a mean value of 101 days; for infants and children, the biological half-life ranges between 5 and 50 days.

### C.2.3  Strontium-90

Strontium-90 ($^{90}$Sr) is not a naturally occurring radionuclide. It has a half-life of 29 years. Almost no gamma ray particles are released from the $^{90}$Sr decay sequence. For example, 99.98 percent of all $^{90}$Sr decays result in the emission of a 523 keV maximum-energy beta particle and a 2280 keV maximum-energy beta particle from $^{90}$Y decay with no gamma ray. Chemically, strontium behaves like the other alkaline-earth metals (e.g., magnesium, calcium, and barium) and has a relatively simple chemistry. In a nuclear reactor fuel element, strontium preferentially forms a stable oxide. When released into the atmosphere, strontium oxide has a high affinity for water and will readily form a soluble hydroxide.

Strontium-90 exists because of the human activities. Due to the high beta particle energies of $^{90}$Sr (and its daughter $^{90}$Y), it has been used in industrial applications to measure thicknesses of paper, plastic, rubber, and metal foils. It also has some medical applications such as treatment for some eye and skin diseases. Due to the low gamma ray activity of $^{90}$Sr, it is not normally used as an irradiation source. The primary means of producing $^{90}$Sr is via nuclear fission, and the main source of $^{90}$Sr is from fission product recovery.

Most of the $^{90}$Sr that exists is trapped inside the spent $UO_2$ fuel in complex chemical compounds. Strontium-90 can be recovered from high-level radioactive waste streams for eventual separate disposal or use in industrial or medical applications. A large inventory of $^{90}$Sr exists at the Hanford Site. Some $^{90}$Sr also exists as Special Case Waste in the form of sealed sources used to

generate known amounts of gamma radiation for thickness gauging and other purposes. Strontium-90 is also used in some radioisotope thermoelectric generators.

The primary low-level waste streams produced at light water reactors include both wet and dry wastes, such as spent-ion exchange resins, filter sludges, filter cartridges, and compactible trash. Most of these wastes will contain small amounts of $^{90}$Sr in the presence of other radioactive nuclides. Small amounts of radioactive waste containing $^{90}$Sr are also generated in research facilities.

Since strontium typically is in a soluble form and chemically bonds less with surrounding soil and rock than many other radioactive species (for example $^{137}$Cs), it tends to migrate further with the ground water. Strontium tends to percolate deeper into the soil due to the effects of leaching by soil moisture. This has the beneficial effect of reducing the probability that strontium will become airborne due to surface erosion, but it also means that strontium can more quickly migrate into an underground water supply. Also because of its solubility, $^{90}$Sr can be taken up by plants through the roots, which is the principal means by which strontium gets into the food chain. Absorption through the upper plant structures (leaves, stem) does not happen to an appreciable degree.

Absorption of strontium by humans takes place primarily through the gastrointestinal tract. Once in the body, up to 95 percent of $^{90}$Sr is eventually excreted within a few weeks. The remaining fraction is fixed within the skeletal structures, preferentially in the areas of bone growth. Health hazards include the possibility of bone cancer. Strontium enters the body and can replace calcium atoms within the bone. The effective half-life of $^{90}$Sr that remains in the skeletal structures is about 18 years (including both decay and biological/chemical processes).

## C.2.4 Uranium

Natural uranium chiefly contains three isotopes of uranium–uranium-234 ($^{234}$U), uranium-235 ($^{235}$U), and uranium-238 ($^{238}$U). Uranium-234 is a member of the $^{238}$U decay chain and usually found in equilibrium with its $^{238}$U parent. The amount of $^{238}$U in natural uranium is more than 99 percent, but the $^{235}$U, present at 0.72 percent in natural uranium, is most radioactive and important in nuclear weapons and nuclear reactors. Enriching uranium, a process by which the percentage of $^{235}$U is increased in relation to the other uranium radionuclides, makes it useful in nuclear weapons and nuclear reactors. Uranium-234 has a half-life of 2.5 x $10^5$ yr and exists as 0.0057 percent of natural uranium. Uranium-230 ($^{230}$U) is also a member of the $^{238}$U decay chain but has a short half-life of only 20.8 days.

Uranium-238 has a half-life of 4.47 x $10^9$ years. This radionuclide decays by alpha particle emission to $^{234}$Th. A series of 14 alpha and beta transitions results in the stable $^{206}$Pb nuclide. Moderately high-energy alpha particles, low-energy x-rays, and low-energy beta particles are emitted during this series of transitions.

The presence of uranium can be very significant in assessing the long-term performance of a LLW disposal facility due to the quantity, radiotoxicity, and mobility of its daughter products, which include isotopes of radium and radon.

Uranium is very reactive and forms compounds with many other elements, such as the halides, oxygen, and hydrogen. The ability of soil to adsorb uranium out of the ground water depends on a number of factors, including pH and the presence or absence of complexing agents. In the

presence of low pH soil conditions, uranium is very soluble and tends to remain in the ground water rather than being adsorbed by the soil.

The principal radiological hazard associated with uranium is due to the relatively high-energy alpha particles its radionuclides and daughters emit.  Since these alpha particles do not penetrate materials easily, external exposure to uranium does not pose a high risk.  The principal risk is due to either inhalation or ingestion.  Inhalation occurs either from release of volatile uranium compound or from suspension of volatile uranium-laden aerosols.  Ingestion can occur when the uranium is introduced into water for consumption or the food chain by plant uptake.  When uranium is either ingested or inhaled, it is removed from the body with a biological half-life varying between 6 and 5,000 days, depending on which organ has become contaminated. Uranium tends to concentrate in the kidneys and the bones.  Additionally, if inhaled, the lungs can receive a dose.

## C.2.5   Plutonium

Plutonium is an element produced in nuclear reactors by neutron irradiation of uranium.  Neutron capture converts uranium-238 ($^{238}$U) into neptunium-239 ($^{239}$Np) which transforms, in a matter of days, into plutonium 239 ($^{239}$Pu).  Absorption of more neutrons and other nuclear reactions generate the other isotopes of plutonium, such as plutonium-238 ($^{238}$PU), plutonium-240 ($^{240}$Pu), plutonium-241 ($^{241}$Pu), and plutonium-242 ($^{242}$Pu).  All isotopes of plutonium are radioactive.

Plutonium-239 is fissile (i.e., fissionable by neutrons), and thus able to sustain a nuclear chain reaction.  This property made $^{239}$Pu a suitable material for nuclear warheads.  Plutonium was produced in special reactors at the Hanford and Savannah River Sites.  No plutonium for weapons has been produced since 1988.

Plutonium-240 is more radioactive and generates more heat than $^{239}$Pu.  In addition to $^{239}$Pu, weapons grade plutonium contains up to 6 percent $^{240}$Pu, fuel-grade plutonium used in breeder reactors contains up to 18 percent $^{240}$Pu, and reactor grade plutonium from power-producing reactors contains up to 24 percent $^{240}$Pu.  Plutonium-238 is intensely radioactive and generates significant quantities of heat.  It is used to make general purpose heat sources and radioisotope thermoelectric generators to produce electricity in spacecraft.

All plutonium isotopes except $^{241}$Pu emit alpha particles as the principal form of radiation. Plutonium isotopes also emit small amounts of gamma and neutron radiation.  Plutonium-241 decays into americium-241 ($^{241}$Am), which is a much stronger source of gamma radiation.

Plutonium is usually stored as metal or as powdered oxide.  Plutonium metal, the form used in nuclear weapons, will corrode to the oxide form if exposed to air or moisture.  Other processes involving plutonium include purification in strong acids, ion exchange, and physical state conversion.  Because of these processes, the forms of plutonium found in the DOE complex include metal, oxide, solutions, and scrap/residues.

Most data on plant uptake of plutonium have shown that the majority of plutonium found in native plants and agricultural crops comes from surface deposition rather than soil-plant absorption via the roots.  Plutonium concentrations are dependant on plant species, plant age, vegetation type, soil pH, positive ion exchange capability, mineral and organic composition, the plutonium chemical form, and duration of the contamination.  Root absorption of plutonium,

when present in soil, does occur; however, translocation to the aboveground portions of the plant is limited to less than 0.01percent of the total plutonium concentration in the soil.

Inhalation of plutonium delivers significant internal radiation doses to the body. Absorption of plutonium via ingestion delivers a much lower internal dose than inhalation. When plutonium enters the body, its biological behavior is determined by its physical and chemical characteristics. Very small plutonium particles are complexed in the blood and deposited in the liver and on bone surfaces. These deposits are metabolized very slowly, with biological elimination half-lives of about 50 to 100 years.

### C.2.6  Carbon-14

Carbon-14 ($^{14}C$) is a long-lived radionuclide with a half-life of 5,730 years. When $^{14}C$ undergoes radioactive decay, the nucleus emits a single beta particle with an energy of 0.156 MeV. There are no radionuclides that decay to form $^{14}C$; therefore, it has no parent. The $^{14}C$ radionuclide is naturally produced in the upper atmosphere by the reaction of neutrons of cosmic ray origin with nitrogen, oxygen, and carbon.

Carbon-14 is also produced in nuclear reactors as a result of absorption of neutrons by nitrogen, carbon, or oxygen present as components of air, coolant, moderator, structural materials, fuel, or impurities. Additionally, $^{14}C$ is produced for preparation of labeled materials used in medical or biological tracer research.

Because of neutron activation of nitrogen-14, an impurity found in nonfuel reactor components, the activated metals waste stream from nuclear reactors contains a substantial amount of $^{14}C$. Additional $^{14}C$ produced in the coolant may add to the contamination of other waste streams including ion exchange resins, concentrated liquids, filter sludge, cartridge filters, and trash.

The chemistry of carbon is quite complex and involves both oxidation-reduction and nonoxidation-reduction reactions. The predominant forms of $^{14}C$ available for transport at a LLW disposal site are carbon dioxide ($^{14}CO_2$), methane ($^{14}CH_4$), carbonate ion ($^{14}CO_3^{-2}$), bicarbonate ion ($H^{14}CO_3^-$), and elemental carbon in activated metals.

The main forms of $^{14}C$ of concern for ground water transport are bicarbonate and carbonate. Bicarbonate is produced during the dissolution of calcium carbonate into water with a pH of 6.5 to 9, or in the dissolving of carbon dioxide. These two categories encompass essentially all of the $^{14}C$ activity in the waste disposal site available for groun-water transport. Ground-water sources should not be contaminated by activated metals with $^{14}C$. The $^{14}C$ found in these metals is expected to remain in the disposal site because $^{14}C$ atoms are dispersed throughout the metal matrix; most of the metals are corrosion resistant (stainless steel); the corrosion products are, in general, insoluble in either freshwater or seawater; and the carbon is in its elemental form and thus unavailable for microbial activity.

Carbon distributes itself quickly among the major environmental components—the atmosphere, the biosphere, and surface waters. Transfer among these components takes place over a period of a few years. Carbon-14 is easily transferred during biological processes and soil-plant interactions that involve carbon compounds.

The metabolism and kinetics of $^{14}C$ in the human body follow those of ordinary carbon. A fraction of carbon introduced into the body is retained as protein, fat, carbohydrates, and other materials. The remainder of ingested carbon is excreted unchanged or is metabolized to $CO_2$, urea, or other metabolites. Inhaled $^{14}CO_2$ rapidly equilibriates with the air in the lung and enters many organic components of body tissue. The ingestion pathway is the primary route for $^{14}C$ incorporation. The corresponding biological half-life of $^{14}C$ is approximately 40 days.

### C.2.7  Technetium-99

Technetium-99 ($^{99}Tc$) is a long-lived radionuclide with a half-life of 213,000 years. When it undergoes radioactive decay, its nucleus emits a single beta particle with a maximum energy of 0.293 MeV. The decay of $^{99}Tc$ forms stable ruthenium-99 ($^{99}Ru$). Oxidation-reduction reactions dominate the chemistry of $^{99}Tc$. The two most common forms of technetium are the pertechnetate ion ($TcO_4^-$) and technetium dioxide ($TcO_2$).

Uranium and plutonium undergoing thermal fission in nuclear reactors produce most of the $^{99}Tc$ that exists as waste. This waste comes in several forms, including ion exchange resins, filter sludge, cartridge filters, and decommissioning waste. Some $^{99}Tc$ is present at certain fuel cycle facilities (such as enrichment facilities) because of the recycling of spent reactor fuel that took place within the DOE complex. Several nuclear powerplant waste streams that may contain $^{99}Tc$ are usually mixed with concrete to create a solid waste form. Some $^{99}Tc$ waste is also generated through medical, industrial, and academic research.

The high solubility of pertechnetate allows it to move quite rapidly in ground-water systems. The migration rate of pertechnetate is expected to be very close to the velocity of ground water unless it is reduced to a less soluble form. The pertechnetate ion is not volatile; its airborne escape from a LLW disposal facility into the atmosphere is not a concern.

The main route of entry of $^{99}Tc$ into the human body is by ingestion. Once in the body, $^{99}Tc$ localizes in the thyroid gland and the gastrointestinal tract. Within 10 hours, it redistributes to the stomach and organs with excretory functions, such as the kidneys and salivary glands. The time required for the body to eliminate one-half of an amount of $^{99}Tc$ by regular processes of excretion is approximately 60 hours. Very little $^{99}Tc$ is assimilated by the muscle or the brain. Hair, however, retains $^{99}Tc$ for long periods of time after a dose and can be a good indicator of $^{99}Tc$ contamination.

### C.3   Processes That Generate LLW

This section describes the major DOE missions that have and are creating LLW within the complex.

### C.3.1  Nuclear Fuel Cycle

Nuclear fuel cycle wastes contain the isotopes of uranium, $^{235}U$ and $^{238}U$, and small amounts of their daughter products. They are produced in the early steps of the fuel cycle, at the conversion facilities, at the enrichment facilities, and at the fuel fabrication facilities.

The gaseous diffusion uranium enrichment complex produces waste where small amounts of uranium are contained in liquids from equipment cleanup that is routed to settling ponds where it precipitates as sludge.

Fabrication of fuel produces LLW in the form of dry solids of $CaF_2$ containing low concentrations of enriched uranium and other low-activity waste. Other uranium-bearing waste are in the forms of liquids and sludges.

### C.3.2  Centrifugation

Centrifugation is a treatment process that removes suspended solids from LLW streams. In a centrifuge, the rapid rotation of a perforated basket or bowl containing the waste stream causes the solids to separate from the liquids by centrifugal action. The liquids are forced out through the basket wall, and the solids collect and are removed by mechanical action or sprays. Centrifuges are used to dewater resins and filter sludges and to concentrate dilute sludges.

### C.3.3  Decontamination and Decommissioning

Decommissioning nuclear facilities at the end of their useful life will produce large volumes of LLW, much of it as contaminated concrete and metal vessels and piping. Decommissioning waste will generally contain the same isotopes as produced during facility operation. Depending on the type of facility and its previous operations, waste streams from the decommissioning may include activated metal, activated concrete, contaminated metal, contaminated concrete, dry solid waste (trash), spent resins, filter cartridges, and evaporator bottoms.

Decontamination technologies involve the removal of deposited radioactivity from contaminated equipment and other solid waste forms by physical and/or chemical methods. They are used at contaminated sites to clean these surfaces so as to restore structures and equipment to a condition to be reused or, prior to demolition, to reduce the potential for demolition waste to be radioactive. Decontamination activities usually involve the use of four processes—physical decontamination, chemical decontamination, electropolishing, and ultrasonic cleaning.

### C.3.4  Remediation

The remediation of radiologically contaminated sites, including buildings, storage piles, equipment, and underlying soils, can be broadly divided into two categories: (1) the decommissioning of nuclear facilities, which may involve the removal of contaminated equipment and structural components and the decontamination of building and equipment surfaces to remove radioactivity; and (2) the cleanup and stabilization of sites that may contain radioactive contamination in buildings, equipment, waste storage piles, and underlying soil. In some instances, the contamination may be detectable as elevated radionuclide levels in ground water, surface water, or airborne pathways.

### C.3.5  Waste Water Treatment

Liquid wastes can be treated to remove contaminants and thus permit the treated liquid to be disposed, or to concentrate the liquid (and contaminants) and reduce the volume to be disposed. Liquids in the form of contaminated ground water or leachate can be pumped to the surface for treatment or can be treated *in situ*. Generally, the treatment of liquids or semisolids will produce

an effluent that can be discharged and a residual sludge or liquid in which the radioactive constituents are concentrated. The processes that follow are often used at radiologically contaminated sites for treatment of waste liquids:

- Chemical precipitation, or separation can be used to remove soluble metals from contaminated ground water or impounded liquids.
- Ion exchange techniques can be used to remove inorganic salts from a liquid waste stream.
- Evaporation can be used to reduce the volume of radioactive liquids and to remove water from a solution.
- Filtration technology can be used to remove suspended solids from a solution by forcing the liquid phase through a porous membrane and collecting the solid phase on the filter medium.
- Activated carbon adsorption is a physical treatment that can be used to remove complex mixtures of organic contaminants from leachate or ground water.

## C.3.6  Stabilization

Stabilization systems are used to immobilize inorganic waste within an inert matrix. Solidification is required for liquid wastes to achieve regulatory limits on water in packages. It is also used for the stabilization of semisolid and solid wastes. There are a number of binder materials commercially available, the selection of which will be dictated by the characteristics of the waste and the relative economics of the competing techniques. Although most waste and soil from a remediated site are shipped offsite or stored in unpackaged bulk form or packaged essentially water free and not stabilized, there are situations in which liquid and/or semisolid wastes will require stabilization and packaging prior to shipment.

Among the available binder materials, cement has the longest record of experience and is applicable to a wide range of waste compositions. The cement is either used as a binder by itself or mixed with a material such as fly ash or cement kiln dust. The waste stream is slurried into this mixture and allowed to set, creating a volume typically twice that of the original waste volume. Thermoplastic binding using asphalt is a competing technique that, although generally more expensive than cementation, provides certain improvements in leaching characteristics and greater volume reduction factors.

## C.3.7  Dewatering

Dewatering is a liquid removal technique using concentration technologies that are particularly applicable to treatment of semisolids. In dewatering, either pumping and/or gravitational drainage is used to remove the water from a semisolid. A commonly employed approach for treating ion exchange resins, called "in-container dewatering," involves the use of multiple filter elements placed in a disposable container and connected to a pump.

### C.3.8  Accelerator Targets

Accelerator targets are used to produce radionuclides through direct interaction with charged particle beams or indirectly through the interaction of induced radionuclides and other materials.

### C.3.9  Weapons Production

Production reactors use high-enriched uranium (HEU) as a fuel to provide fission neutrons for capture by depleted uranium (DU).  Uranium-238 captures the neutron to form $^{239}$U, which decays to $^{239}$Pu.  If the target material is left in the process too long, an increased amount of $^{240,241}$Pu will be formed.  Production reactors also produce tritium.  The $^{235}$U fission produces neutrons that are captured by a lithium-6 target, resulting in an alpha emission and tritium ($^{3}$H).  Other major isotopes produced in a production reactor include $^{238}$Pu, $^{237}$Np, $^{252}$Cf, $^{241}$Am, $^{60}$Co, $^{137}$Cs, and $^{85}$Kr.

There are various reprocessing methods to recover special nuclear materials from irradiated fuel and targets; the method used depends on the material the fuel is made from.  All these methods, however, use some form of the Purex process.  Purex is a solvent extraction system that uses tributyl phosphate to separate the materials in the fuel into transuranic elements, uranium, plutonium, and fission products.  The fission products fraction contains the waste elements, and they are highly radioactive.  These liquid wastes are evaporated and stored in underground tanks until decay heat has been reduced sufficiently for preparation for ultimate disposal.

### C.3.10 Ion Exchange

Ion exchange is a process involving the selective removal of contaminants from liquids through the reversible interchange of similarly charged ions between an electrolyte solution and a solid phase.  The contaminants are accumulated on to the exchange medium, which is typically a resin.  Ion exchangers are either of the cation or anion type.  A further categorization is between separate-bed systems (demineralizers) consisting of either cation or anion resins, and the mixed-bed system consisting of a stationary bed containing fixed anion and cation resins.  The mixed-bed systems have been the predominant ion exchange choice for treatment of liquid wastes.

### C.3.11 Nuclear Reactors

Control of radioactive materials in a power reactor is generally achieved by removing material from process streams, concentrating it in a relatively small volume, and disposing of that volume as LLW.  Small amounts of radioactive material are present in the coolant of a power reactor from fuel leakage as well as from system corrosion products and impurities in the coolant that have been activated by neutron bombardment.  The radioactive waste treatment systems are designed to remove these materials on an ongoing basis through filtration and ion exchange resins in both the primary system and secondary, or auxiliary, systems that treat liquids with which the primary coolant may have come in contact.

Some of the LLW generated during the operation of a nuclear reactor include:

- ion exchange resins;
- filters that remove particulates from liquids or from air;
- concentrated liquids resulting from evaporative processes;

- dry active waste, which is a term generally applied to a wide variety of waste products such as cleaning materials, glass, filters, concrete, miscellaneous wood, and metals;
- nonfuel reactor components, such as fuel channels, control rods, and in-core instrumentation; and
- decontamination wastes.

### C.3.12 Research Facilities

Some DOE facilities use radioactive materials in research and employ a variety of radioisotopes and material forms to achieve their purposes. Several waste streams are generated as a result of research and development activities: liquid scintillation vials, other organic and inorganic liquids, biological wastes, and trash. Accelerator targets and sealed sources are also produced. Certain research and development activities concerned with the fuel cycle processes or weapons production will generate waste streams similar to those processes.

### C.3.13 Isotope Production

Processes that result in the generation of LLW include the production and distribution of radioisotopes for medical, academic, or industrial use; manufacture of materials containing radioisotopes; and the use of radioisotopes for research and testing and in gauges and instrumentation. Several distinct waste streams vary in the concentration of activity, isotopes contained, and volumes produced. Isotope production is achieved through irradiation of fuel and targets in a reactor, and the separation and purification of the resulting products. LLW generated from medical isotope production includes solidified aqueous liquids and trash produced in the separation, cleanup, and shipping of the radioisotopes.

### C.4    Special Case Waste

Special case waste is waste that does not meet the waste acceptance criteria (WAC) of any of the DOE disposal facilities. Usually, this is because the waste

- is too big and cannot be reduced in size,
- has radionuclide concentrations that exceed site-specific limits,
- contains hazardous waste prohibited by the WAC, and/or
- is classified for national security purposes.

The following categories of special case waste currently exist within the DOE complex. Because of the general nature of the category definitions, some overlap exists. For the purposes of this document, the special case category "Mixed Waste" was excluded. Estimated volumes and radioactivity totals are presented in parentheses after each category title (Sandia National Laboratories 1994).

### C.4.1   Non-Certifiable Defense Transuranic (TRU) Wastes

This category includes TRU wastes generated through the defense program that are not certifiable for disposal at WIPP or for transport in currently used shipping containers. TRU wastes generally result from reprocessing spent fuel and from fabricating plutonium weapons and plutonium-bearing reactor fuel. Examples of non-certifiable defense TRU wastes include:

- some wastes from decontamination and decommissioning of hot cells,
- large metallic structures containing or contaminated with TRU elements,
- large pieces of alpha- or fission-product contaminated equipment that require remote handling,
- nuclear weapons accident residue, and
- classified defense-generated TRU waste.

## C.4.2  Non-Defense TRU Wastes

This category includes DOE-titled wastes generated by DOE Energy Research/Nuclear Energy Programs or an NRC licensee.  Some of these wastes also contain mixed activation products or mixed fission products in addition to TRU isotopes.  Examples of non-defense TRU wastes include:

- HEPA filters from hot cells,
- dewatered ion exchange resins,
- cartridge filters from nuclear reactors,
- neutron sources containing TRU isotopes,
- reactor vessels,
- submerged demineralizer systems, and
- wastes generated from decontamination and decommissioning of fuel fabrication facilities.

## C.4.3  Specific Performance Assessment-Required Wastes

This category includes DOE-titled LLW with radionuclide concentrations that exceed the limits for Class C waste given in 10 CFR 61.55.  This waste is similar to non-defense TRU waste, except that specific performance assessment-required wastes do not have concentrations of TRU in excess of 100 nCi/g.  Examples of specific performance assessment required wastes include:

- $^{90}$Sr radioisotope thermoelectric generators,
- Three Mile Island-2 submerged demineralizer system liners,
- hot cell wastes,
- equipment contaminated with alpha and mixed fission products,
- ion exchange resins,
- sludge, and
- filter media that contain uranium and $^{99}$Tc.

## C.4.4  Performance Assessment Limiting Wastes

This category includes DOE-titled LLW with radionuclide concentrations that exceed site-specific performance assessment limits for disposal, but do not exceed the limits for Class C waste as given in 10 CFR 61.55.  Examples of performance assessment-limiting wastes include:

- absorbed tritiated liquids,
- hot cell wastes from destructive examination of fuels,
- sludge containing mixed fission products,
- ion exchange resins containing TRU,
- gauges and dials containing $^{226}$Ra, and
- uranium solids with associated decay products.

### C.4.5 Fuel and Fuel Debris Wastes

This category includes DOE-titled fuel and fuel debris wastes used for research and development purposes. These wastes are similar to wastes destined for the HLW repository; however, they are not in typical HLW packaging configurations and may not meet the HLW repository WAC. Most of the activity comes from mixed fission products, but TRU isotopes are also present. Examples of fuel and fuel debris wastes include:

• material from the core of the Three Mile Island-2 reactor,
• fuel from various DOE test reactors, and
• fuel and debris from various DOE research and development projects.

### C.4.6 Uncharacterized Wastes

This category includes containers of waste with unknown contents. These containers may hold nuclear materials at or near the limits of greater-than-Class-C or TRU wastes. Any of the uncharacterized waste that is found to be special case can most likely be placed in one of the other special case waste categories after characterization. The primary example of uncharacterized waste is the underground tanks at the Hanford Site.

### C.4.7 Excess Nuclear Materials Wastes

This category includes nuclear materials above the economic discard limit that becomes waste because they cannot be reprocessed to recover the special nuclear material. These materials are either no longer useful or require processing that cannot be performed because of capability or capacity constraints of present processing facilities. Examples of excess nuclear materials wastes include:

• unirradiated nuclear materials,
• irradiated nuclear materials,
• nuclear materials containing decay products,
• six-foot diameter metal spheres that contain plutonium isotopes,
• $UF_6$ gas cylinders from isotope separation research, and
• items containing significant quantities of fissionable and fissile nuclear material.

### C.4.8 Sealed Sources

This category includes encapsulated radioactive material whose main purpose is to generate known amounts of gamma radiation, heat, or neutrons. Radionuclide concentrations usually make these sources greater-than-Class-C or TRU wastes for disposal purposes. Over 30 different radionuclides are used to make sealed sources, but the ones most commonly used are $^{60}$Co, $^{90}$Sr, $^{137}$Cs, $^{238}$Pu, $^{239}$Pu, $^{241}$Am, $^{244}$Cm, and $^{252}$Cf. Other isotopes include $^{75}$Se, $^{124}$Sb, $^{147}$Pm, and $^{204}$Tl. DOE is the largest supplier of sealed sources, and NRC licensees have received sealed sources from DOE under a variety of contractual agreements. Those sealed sources that do not have to be disposed of in an NRC-licensed facility, and that have radionuclide concentrations exceeding Class C limits, are considered special case waste.

### C.4.9   DOE-Titled Wastes Held by NRC Licensees

This category includes DOE-titled wastes or materials that are held by NRC licensees under a contract, loan, lease, or grant for use in nuclear research-related fields.  These are considered special case wastes when the pedigree of ownership is in question.  Examples of DOE-titled wastes held by NRC licensees include:

- sealed sources,
- uranium slugs,
- uranium fuel elements,
- thorium,
- heavy water, and
- fission chambers.